

REMARKS

Reconsideration of this application is requested. Claims 1-3 and 5-16 will be active in the application subsequent to entry of this Amendment.

The sole issue raised in the outstanding Official Action is the patentability of claims 1-16, which the examiner has denied on the basis of the disclosures of a single document. Applicants appreciate the detailed discussion provided by the examiner of the content of the document relied upon, however they are confident that the claims of this application, as above amended, define subject matter that is both novel and inventive over the disclosure of the applied reference.

Claim 1 has been amended in order to more particularly point out and distinctly claim that which applicants regard as their invention and to direct the claim to a preferred aspect of the disclosure.

Claim 1 is revised and directed to the production of methacrylate-based block copolymers as described in the specification generally at page 6, lines 3-7, also the embodiment of claims 7-16 at page 7, line 12 to page 8, line 26, and in the working examples.

The process includes the second polymerization of monomer containing methacrylate monomer (the first polymerization is the preparation of a macroinitiator). Applicants address a difficulty of this second polymerization in the conventional methods and have found a novel and inventive way to solve this art-recognized difficulty.

All claims are rejected as being anticipated by or obvious over the disclosure of Matyjaszewski et al US 6,407,187 (hereinafter '187). Applicants' claims include the following features:

- (1) Molar ratio of  $(M)^n$  to  $(M)^{n+1}$  of 90/10 to 0.1/99.9
- (2) In the second polymerization, methacrylate-based of monomer is polymerized, not disclosed in '187.

The differences between the invention claims 7 and 13 and '187 are the same difference as described above.

As described in the Office Action, page 4, lines 1-6, '187 at column 14, states that a block copolymer can be obtained by living polymerization. Further, in '187, column 17, the patent reports a copolymer having narrow molecular distribution ( $M_w/M_n \leq 1.5$ ) can be obtained. However, in '187 there is no description nor suggestion that a block copolymer is produced by polymerizing a methacrylate-based monomer in the second polymerization, the process to which applicants' claims are directed.

In '187's Examples 12, 18 and 22-23 (columns 37-39), polymers having narrow molecular distribution ( $M_w/M_n$ ) were obtained using methacrylate as a monomer and low-valence metal complex as a catalyst under different conditions from that as defined in the amended claim 1. However, in '187, there is no description nor suggestion that a block copolymer is produced by polymerizing a methacrylate-based monomer in the second polymerization as summarized below.

Example No.	12	18	22	23
Polymer type Produced	PMMA (homopolymer)	P (St-co-MMA) (Random copolymer)	PMMA (telechelic homopolymer)	Modified PMMA (Ex 22)
Solvent	Ethyl acetate	-	Ethyl acetate	-
Polymerization time (hr)	1.4	14	8	-
MN	20,500	38,000	5,000	-
Mw/Mn	1.40	1.55	1.35	-

From the above summary of the data in representative examples of '187, there is no description nor suggestion of a technical object to be solved by the present invention, that is, providing a convenient and facile second polymerization of monomer containing a methacrylate monomer which is difficult to conduct using conventional methods. Further, '187 provides no description nor suggestion of this technical problem nor that it can be solved by using a specific solvent and metal complex catalyst whose molar ratio of low-valence metal to high valence-metal is defined as the above specific range.

Applicants address the difficulty of producing a block copolymer including the second polymerization of monomer containing methacrylate monomer present in conventional methods. By the present invention, this difficulty can be solved, that is, a

methacrylate-based block copolymer having narrow molecular distribution can be produced.

Applicants' investigations have been directed to solving a long-standing problem in the art. Applicants have found when applying the atom transfer radical polymerization method to the production of block copolymers, the block polymerization sometimes fails to proceed smoothly depending upon kinds of monomers used. It is thought that the poor block polymerization is caused by the difference between ease of abstracting the halogen atom from the end of the polymerization initiator and ease of abstracting halogen bonded to the end of the successively added monomer, i.e.,  $K_{act}$  in the reaction formula shown on page 2 of applicants' specification. In particular, in the case where the first block chain is constituted from acrylate-based monomers, since the rate for abstracting halogen from the end of the acrylate is too slow, it becomes difficult to conduct the subsequent formation (i.e., block polymerization) of the second block chain composed of methacrylate-based monomers.

The advantages provided by the present invention are demonstrated at page 54, line 8 (under Table) to page 61, line 9 and Page 61, line 18-27 (corresponding to the inventions of claims 1 and 7).

The process in Example 9 using macroinitiator A produced in Reference Example 1 and a redox catalyst (halogen: chlorine) provides more excellent results than that of Examples 10 and 11 using macroinitiator A produced in Reference Example 1 and a redox catalyst (halogen: bromine), in attaining narrow molecular weight and degree of halogen in the end group.

The process in Example 12 using macroinitiator B produced in Reference Example 2 and a redox catalyst is superior to that of Example 13 using macroinitiator B produced in Reference Example 1 and a redox catalyst (using only low-valence metal catalyst), in attaining narrow molecular weight and degree of halogen in the end group.

To summarize, Examples 1 to 13 confirm that when the methacrylate-based monomer was polymerized in the presence of the redox catalyst having a molar ratio of

the low-valence metal  $(M)^n$  to the high-valence metal  $(M)^{n+1}$  of 90/10 to 0.1/99.9 upon initiation of the polymerization, it was possible to produce the methacrylate-based polymer having a narrow molecular weight distribution. Also, as shown in these Examples, it was confirmed that when the methacrylate-based block chain was initiated in the presence of the redox catalyst having a molar ratio of the low-valence metal  $(M)^n$  to the high-valence metal  $(M)^{n+1}$  of 90/10 to 0.1/99.9 subsequent to the step for forming the acrylate-based block chain, it is possible to produce a block copolymer having a narrow molecular weight distribution and containing both the acrylate-based block chain and the methacrylate-based block chain.

The data contained in applicants' as-filed specification provides the type of illustration/showing contemplated by the examiner on page 5, lines 1-2 of the Official Action. The results presented in the original specification accompanied by the executed declaration signed by the inventors have significant evidentiary weight, comparable to the weight given to an executed declaration. It is well established by the Federal Circuit that "the examiner must consider comparative data presented in the specification which is intended to illustrate the claimed invention in reaching a conclusion in regard to the obviousness of claims." *In re Margolis*, 785 F.2d 1029, 228 U.S.P.Q. 1123, 1129 (Fed. Cir. 1993).

Page 62, line 1 to page 67, Table 14 shows the results corresponding to the invention of claim 13.

The process in Examples 14 and 15 using macroinitiator C and D produced in Reference Examples 3 and 4, respectively, are more excellent than that of Example 16 using macroinitiator C produced in Reference Example 3, in attaining narrow molecular weight.

For the above reasons it is respectfully submitted that the claims of this application define inventive subject matter. Reconsideration and allowance are solicited.

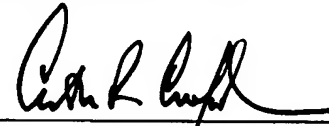
SAITO et al  
Serial No. 10/023,220

Attached hereto is a marked-up version of the changes made to the claims by the current amendment. The attached page(s) is captioned "Version With Markings To Show Changes Made."

Respectfully submitted,

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VERSION WITH MARKINGS TO SHOW CHANGES MADE

IN THE CLAIMS

1. (Amended) A process for producing a methacrylate-based [polymer] block copolymer, comprising:

polymerizing (d1) a radical-polymerizable monomer containing at least one methacrylate-based monomer in the presence of (c1) a redox catalyst comprising a metal complex containing at least one transition metal as a central metal selected from the group consisting of elements of Groups 7 to 11 of the Periodic Table, said redox catalyst containing a low-valence metal  $(M)^n$  wherein  $n$  represents an atomic valence of the metal, and a high-valence metal  $(M)^{n+1}$  both constituting the redox catalyst system, and having a molar ratio of  $(M)^n$  to  $(M)^{n+1}$  of 90/10 to 0.1/99.9, upon initiation of the polymerization, using (a1) at least one polymerization solvent selected from the group consisting of water, ethers, amides, nitriles and alcohols, and (b1) a polymerization initiator selected from [the group consisting of organohalogen compounds, halogenated sulfonyl compounds and] halogen-containing macroinitiators.